

Argonne National Laboratory

PILOT-LOT FABRICATION OF
ZPPR OXIDE-ROD ELEMENTS

by

J. E. Ayer, A. G. Hins,
and F. D. McCuaig

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Materials Science Division

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PILOT-LOT FABRICATION OF ZPPR OXIDE-ROD ELEMENTS

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ABSTRACT

The development and manufacture of oxide-rod fuel elements and gamma-scan standards for the ZPPR reactor are discussed. A total of 52 elements: 30 oxide-rod fuel elements and 22 gamma-scan elements, were made. All UO_2 and $(\text{U,Pu})\text{O}_2$ pellets were encapsulated in Type 304L stainless steel jackets, with helium at atmospheric pressure.

The UO_2 and $(\text{U,Pu})\text{O}_2$ pellets were fabricated by the Dow Chemical Co., Rocky Flats Division, and sent to Argonne National Laboratory. The pellets were made by a cold-press and sinter technique that produced acceptable pellets without centerless grinding. The pellets, produced with a length-to-diameter ratio of 1.5, had a theoretical density of about 95%.

A loading technique was developed to protect the outside of the jacket tube and weld zone from plutonium contamination. A special end plug was designed to prevent thinning of the jacket tube wall or an increase of the outside diameter in the welded area, and to produce a finished element of a predetermined overall length. The elements were dimensionally checked, X-rayed, leak detected, and surveyed for alpha contamination on the external surfaces before final acceptance.

I. INTRODUCTION

Data from the fabrication of a small number of UO_2 and $(\text{U,Pu})\text{O}_2$ right-circular cylindrical pellets were used, in conjunction with a literature survey, as a basis for a product specification governing the fabrication of fuel elements for ZPPR. Exact diameters of the pellets were not specified; instead, weights of material that could be introduced into a jacket with an inside diameter of 0.350 in. and a nominal length of 5.7 in. were fixed. This effectively required the diameters to be between 0.340 and 0.345 in., with a theoretical density of at least 92%. The objective of this requirement was to

permit sufficient latitude on fired dimensions and densities to preclude centerless grinding. Two compositions of $(U,Pu)O_2$ were required: $(U_{0.85}Pu_{0.15})O_{2-x}$ and $(U_{0.70}Pu_{0.30})O_{2-x}$. The substoichiometry was desired because it is conveniently attained in reducing atmospheres and the mixture is single phase at equilibrium.¹

In addition to writing a product specification and obtaining statistical information, loading studies were conducted with part of the pellets that were produced. The loading studies were required to prove adequacy of design as well as to identify and solve any major problems associated with pilot production of prototypic elements. At least two problems associated with fuel-element assembly were anticipated. The first of these was the control of plutonium contamination on the outside of the fuel-element jacket, particularly in the area of the weld. Contamination of the weld area cannot be tolerated, since radioactive particles will be trapped in the fusion zone and will result in a "count" that is difficult, if not impossible, to remove. The second problem area was related to the maintenance of minimum diametral tolerances between fuel and jacket and between jacket and the calandrias that will contain the fuel elements during the reactor physics experiments. Reactor physics considerations established radial clearances and dimensions as: fuel-to-jacket, 0.003 in.; clad thickness, 0.012 in.; and clad-to-calandria, 0.006 in. The small gap between the jacket and the plutonium-bearing fuel aggravates the contamination problem and loading difficulties. The limited gap between the clad and the calandria means that weld-bead protrusion must be kept at a minimum.

II. DESIGN OF THE FUEL ELEMENT

The geometric characteristics of the ZPPR oxide-rod element are shown in Fig. 1. Of prime importance is the straightness tolerance designated as 0.002 in. This dimension limits the rod straightness and

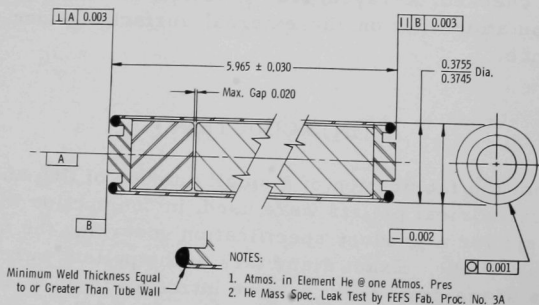


Fig. 1. ZPPR Oxide-rod Element

weld buildup to a total of 0.002 in., assuring adequate clearance between the element and the calandria tubes. The $+0.000 - 0.020$ in. gap between the fuel and the end cap is maintained by interposing a spacer of Type 304 stainless steel between the fuel column and end cap. The length of the spacer is determined for each individual element. The fuel-element length tolerance of ± 0.030 in. results from the algebraic accumulation of tolerances on the fuel tube and the two end plugs, which are ± 0.005 and ± 0.010 in., respectively. Figure 2 depicts the fuel pin. The pin consists of pellets that

were produced by the Dow Chemical Co., Rocky Flats Division, as output from a program to evaluate manufacturing processes for UO_2 and $(\text{U,Pu})\text{O}_2$ fuel-rod elements.

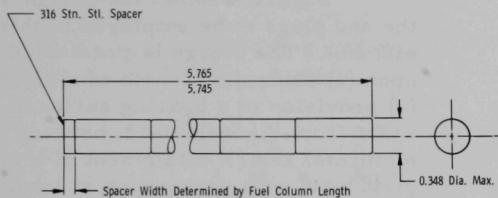


Fig. 2. ZPPR Oxide Fuel Pin. Neg. No. 52202.

MIL-STD-8C.* The choice of jacket material is Type 304L stainless steel that conforms to ASTM designation A269-65. Type 304L stainless steel is a commonly available reactor-grade tubing for containment of $(\text{U,Pu})\text{O}_2$ fuel. The intent of the physics experimenter to heat the subject fuel elements to 600°C repeatedly and their right to expect little, if any, degradation of clad integrity in the process prompted the choice of this alloy. Work by Ruther and Greenberg² and by Gulbransen and Andrew³ shows that at 600°C the rate of weight change of Type 304 stainless steel in purified oxygen at a pressure of 76 mm Hg was linear with time between 2 and 6 hr. A weight-gain rate of about $0.2 \mu\text{g}/\text{cm}^2/\text{hr}$ was found for this condition. This weight gain, calculated for a 0.375-in. tube 6 in. long is 3.6×10^{-6} g/hr, which is a low corrosion rate. It is significant that the oxide film or scale formed on Type 304 stainless steel is adherent and resists spalling during cooling.

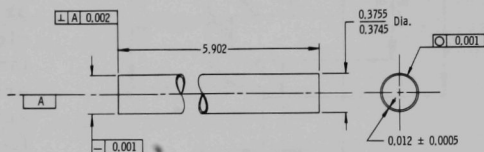
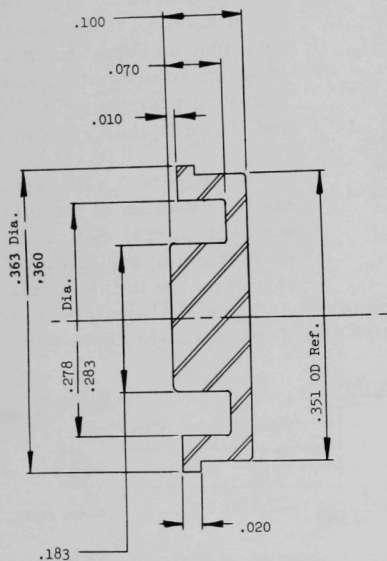


Fig. 3. ZPPR Oxide-fuel Tube. Neg. No. 52203.

The outside diameter of the tubing was specified as 0.3750 ± 0.0005 in. and the thickness as 0.0120 ± 0.0005 in. These conditions yield an inside diameter of 0.3495 in. minimum and 0.3525 in. maximum and, correspondingly, a fuel-jacket gap of from 0.0022 to 0.0038 in. with a 0.3450-in.-OD fuel. The roundness of the tubing, that is, the difference between the major and minor

*USASI Y14.5, 1966.

axis length of an oblate tube, is set at a maximum of 0.001 in. This dimension is consistent with the diameter tolerance and is set at this level to insure sufficient inside clearance to permit pellet loading. The tubing straightness tolerance of 0.001 in. is required to control the jacket-calandria gap and to provide some allowance for weld-bead protrusion. The perpendicularity tolerance between the tube centerline and the face of the tube limits the gap between tube face and end plug, and ensures a uniform surface against which the end closure weld is made.



NOTE:

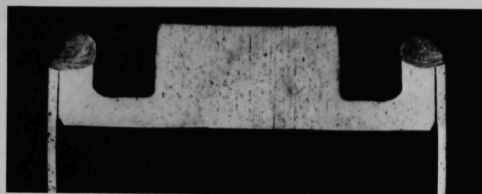
1. .351 Dia. To Have a Class LC₂ Fit with Tubing ID (Ref. Machinery's Handbook, 17 Edition p. 1532)

Fig. 4. ZPPR Oxide-rod End Plug. Neg. No. 52204.

Figure 4 shows the design of the end plugs to be employed in this element. The design is predicated upon (1) controlled length of element, (2) provision of a bearing surface, apart from the weld bead, between elements, and (3) attainment of a girth weld with penetration at least equal to the wall thickness of the tubing with minimum protrusion beyond the outside diameter of the element. These criteria are met in the following manner. The 0.010-in. protrusion over the end-plug flange provides a surface against which elements may be abutted in the calandria without contacting the weld area. The flange on the end plug serves a number of purposes. The flange provides a seat for the end cap on the tube face permitting overall length control; seating establishes constant plug location against the force applied by chills to insure intimate contact; and provides sufficient metal to achieve full tube-thickness weld penetration without significant bead buildup on the weld

periphery. The undercut in the end plug serves two purposes. It establishes the effective weld thickness which, in turn, determines the amount of metal in the weld bead. It also provides a well to accommodate a chill that, when properly placed, keeps the weld bead from running to the inside to the extent that it thins the outer tube wall.

Figure 5 is a photomicrograph of a typical weld made against the tube and the end-plug design shown in Figs. 3 and 4, respectively.



Finished Weld
Magnification: $\sim 7.5X$

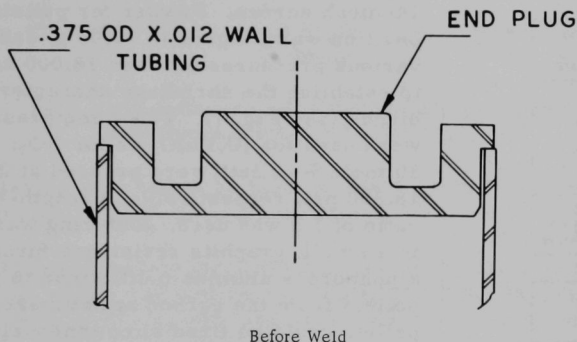


Fig. 5. ZPPR Oxide-rod Weld Configuration,
ANL Neg. No. 350-1078.

III. PELLET MANUFACTURE⁴

A. Experimental Procedure

The manufacture of the $(U,Pu)O_2$ compositions is discussed in this section. Since the fabrication of the UO_2 pellets is inherently simpler, reference to that fabrication will be limited to results.

Two compositions of $(U,Pu)O_2$ were made by mechanical mixing and fabricated according to the schematic in Fig. 6. Hyperstoichiometric UO_2 was obtained from Nuclear Fuel Services through ANL. The plutonium dioxide was made at Rocky Flats by calcining $(Pu\ IV)$ oxalate at $650^\circ C$. A list of impurities and powder characteristics for both oxides is given in Table I. The boron and silicon analyses in the PuO_2 are inordinately high and cannot be explained. The analyses of the particle size and the surface area of the PuO_2 are somewhat surprising since the calcining temperature of $650^\circ C$ should have produced a product of much higher surface area.⁵

In general, it is advisable to attempt to match surface areas of the two oxides being blended. Because of the large PuO_2 agglomerate size it

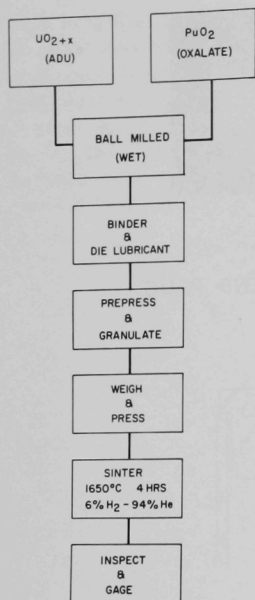


Fig. 6. Schematic of Fabrication
Used for Mixed Oxides.⁴
Neg. No. 51835.

TABLE I. Analysis of UO₂ (Depleted) and PuO₂

Impurity	UO ₂ , ppm	PuO ₂ , ppm	Impurity	UO ₂ , ppm	PuO ₂ , ppm
Ag	<0.10	<2.5	Mn	<5	7
Al	4	67	Mo	<1	3
Am	-	1140	Na	<50	-
As	-	<2.5	Ni	52	63
Au	-	<5	P	<10	<500
B	<0.20	250	Pb	<1	5
Ba	-	2	Pd	-	2.5
Be	<0.1	<0.05	Si	80	449
Ca	<20	-	Sn	-	<3
Cd	<0.10	<2.5	Sr	-	<2.5
Co	<5	-	Th	<100	-
Cr	10	56	Ti	<1.0	2.5
Cu	<3	3	V	<2.5	<2.5
Du	<0.02	-	Zn	30	<10
Eu	<0.02	-	Zr	4.0	-
Fe	48	367	C	63	-
Ga	-	2	N	<10	Average particle size UO ₂ -0.60 μm
Gd	<0.01	-	F	<10	
In	-	<2.5	H ₂ O	0.10	
K	-	<2.5	% U	87.56	
Li	<50	-	O/U	2.11	
Mg	9	<2.5			

Particle Size Analysis PuO₂

3 μm--56%
3-5 μm--17%
5-10 μm--21%
>10 μm-- 6%
Largest--25 μm

Isotopic Analysis, %

238Pu-- 0.035
239Pu--86.303
240Pu--11.468
241Pu-- 1.990
242Pu-- 0.204

was ball milled in water for 24 hr in a rubber-lined mill. Alumina balls were used as the grinding media. An equal weight of UO₂ powder was then added and milled for 4 hr. Additional UO₂ was added to adjust to the required composition and milled for another 4 hr. After removal of the water, 1 wt % Carbowax 4000 and 1 wt % stearic acid were added in a CCl₄ solution. After evaporation of the CCl₄, the mixtures were isostatically pressed at 10,000 psi and granulated through a 14-mesh screen. Powder for pellets of each composition was weighed to ± 0.05 g, and pressed at various pressures between 18,000 and 40,000 psi to establish the shrinkage characteristics of the blends (see Fig. 7). The same press and dies were used for (U,Pu)O₂ as for UO₂. The 15 and 30 mole % pellets were pressed at 20,000 and 18,250 psi, respectively. A length-to-diameter ratio of 1.5 was used. Sintering was accomplished in a small, graphite resistance furnace fitted with a nonporous alumina muffle tube to protect the pellets from the carbon atmosphere. Thirty-eight pellets could be fired simultaneously in an alumina boat. Ten-mesh alumina grain was used for bedding the pellets. Nominal sintering parameters were 1650°C for 4 hr in a dry 94% He-6% H₂ atmosphere. A 16-hr sintering schedule was used. As with the

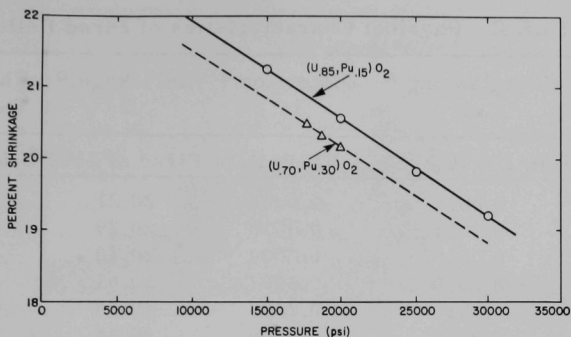


Fig. 7. Shrinkage Characteristics of Two Compositions of Mixed Oxides Sintered at 1650°C.⁴ Neg. No. 51873.

UO₂, the sintering rate had to be held to less than 100°C/hr above 1200°C in order to maintain good dimensional control.

The sintered pellets were visually inspected for defects and measured at the top, waist, and bottom diameters with a blade micrometer. Five pellets from each sintering run were randomly selected for density determinations. Densities were obtained using monobromobenzene as the immersion liquid. Plutonium concentrations were determined by a calorimetric method. Oxygen-to-metal (O/M) ratios were determined by a standard oxidation-reduction cycle.⁶ Homogeneity was examined with the use of an electron microanalyzer monitoring Pu-M β and U-M α X-ray lines and standard techniques of X-ray diffraction.

B. Results

Table II lists the results on hourglassing (maximum diameter minus waist diameter), distortion (bottom diameter minus top diameter), shrinkage, and theoretical density for both (U,Pu)O₂ compositions. The hourglassing and distortion data are very similar to those obtained with pure UO₂. It is apparent from the data that the densities are not affected by the PuO₂ concentration.

The pellets listed in the last section of Table II were a repeat series of those shown in the middle section of the table. The latter were lost due to severe surface spalling, presumably due to packaging. Harvey, Teter, and Leggett postulated that a small amount of carbide formed on these pellets during the sintering operation. This was caused by a crack in the alumina muffle tube, which was later observed. The spalling occurred because of a water-carbide reaction between the pellets and moisture absorbed onto cotton packing material that surrounded each pellet during shipment. Runfors has observed a similar effect on UO₂ sintered in a graphite tube furnace.⁷

TABLE II. Physical Characteristics of Fired Pellets⁴

Run No.	Hourglassing, ^a in.	Distortion, ^a in.	Shrinkage, ^a %	Theoretical Density, %
Data from 296 ($U_{0.70}Pu_{0.30}$)O ₂ Pellets Fired at 1660°C for 5 hr				
5/17/68	0.0026	0.0007	20.21	95.51 ^b
5/20/68	0.0053	0.0010	20.29	93.90 ^b
5/21/68	0.0038	0.0009	20.40	93.68 ^b
5/22/68	0.0030	0.0007	20.61	94.51 ^b
5/23/68	0.0031	0.0007	20.64	95.06 ^b
5/24/68	0.0030	0.0006	20.54	94.59 ^b
5/25/68	0.0023	0.0005	20.54	94.17 ^b
5/27/68	0.0023	0.0007	20.56	93.69 ^b
Data from 260 ($U_{0.85}Pu_{0.15}$)O ₂ Pellets Fired at 1660°C for 5 hr				
4/23/68	0.0010	0.0004	20.50	94.04 ^c
5/6/68	0.0032	0.0007	20.61	95.11 ^c
5/7/68	0.0037	0.0006	20.60	94.86 ^c
5/8/68	0.0056	0.0006	20.50	95.41 ^c
5/9/68	0.0046	0.0007	20.67	96.11 ^c
5/10/68	0.0036	0.0008	20.75	95.84 ^c
5/14/68	0.0021	0.0007	20.45	94.42 ^c
5/15/68	0.0025	0.0007	20.77	95.64 ^c
Data from 240 ($U_{0.85}Pu_{0.15}$)O ₂ Pellets Fired at 1650°C for 4 hr				
6/17/68	0.0049	0.0006	20.60	95.22 ^d
6/18/68	0.0031	0.0006	20.60	95.00 ^d
6/19/68	0.0024	0.0004	20.70	94.81 ^d
6/20/68	0.0058	0.0005	21.06	95.60 ^d
6/21/68	0.0052	0.0007	21.12	95.81 ^d
6/22/68	0.0035	0.0008	21.30	95.69 ^d

^aAveraged from all pellets in run.

^bAverage of 5 pellets randomly selected from each run. Overall average was 94.36%. Standard deviation was 1.00%.

^cAverage of 5 pellets randomly selected from each run. Overall average was 95.31%. Standard deviation was 0.81%.

^dAverage of 5 pellets randomly selected from each run. Overall average was 95.34%. Standard deviation was 0.55%.

Figures 8-10 show the diametral variation for the three batches of pellets. The diametral variation is slightly larger than was observed with UO₂, probably due to the fact that only 38 pellets could be sintered at one time and sintering conditions could not be reproduced exactly.

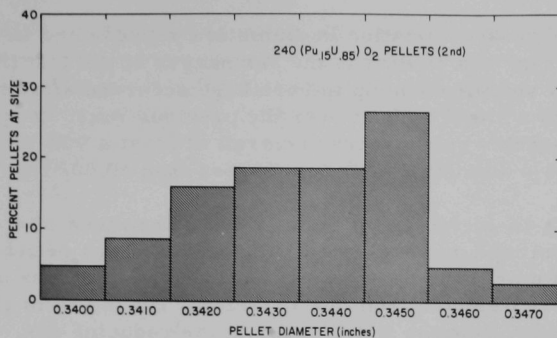


Fig. 8. Histogram⁴ of Pellet Diameters of Mixed
(Pu_{0.30}U_{0.70})O₂. Neg. No. 51875.

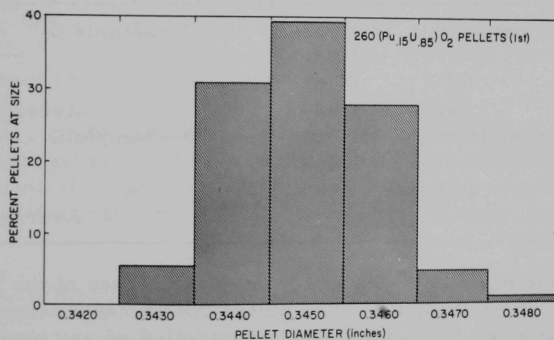


Fig. 9. Histogram⁴ of Pellet Diameters of Mixed
(Pu_{0.15}U_{0.85})O₂. Neg. No. 51874.

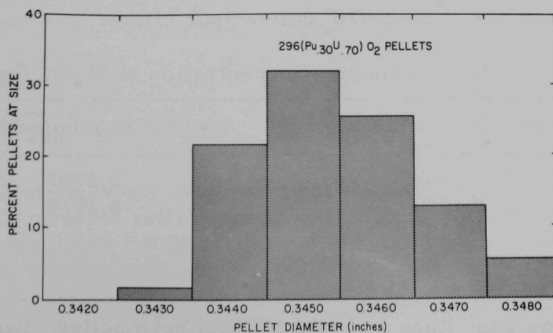


Fig. 10. Histogram⁴ of Pellet Diameters of Mixed
(Pu_{0.15}U_{0.85})O₂. Neg. No. 51876.

The increased variation in diameters represented in Fig. 10 compared with those in Fig. 9 is evident in the shrinkages of the last three runs shown in Table II. A significant jump in shrinkage occurred after the gas flow had been increased a small amount over the previous runs. It appears, however, that with reasonably good process control, at least a 90% yield can be obtained on pellets with a diametral variation of less than ± 0.002 in.

Oxygen-to-metal (O/M) ratios were determined on a random sampling of both compositions and are listed in Table III. Each pellet was crushed and divided into two or three parts and sampled. The averages are somewhat lower than were expected from the literature, but the ratio can be easily adjusted by controlling the water vapor in the reducing gas.

TABLE III. O/M Ratios of Mixed Oxides⁴

wt % PuO ₂ (nominal)	O/M	wt % PuO ₂ (nominal)	O/M
15--Sample A	1.965	30--Sample C	1.943
	1.950		1.943
	1.965		1.942
Average	1.959	Average	1.943
15--Sample B	1.921	30--Sample D	1.960
	1.967		1.950
Average	1.944		1.945
		Average	1.952

The plutonium contents and impurity analyses of the two compositions are listed in Tables IV and V. The plutonium concentrations are slightly higher than expected but can be easily controlled on any production process. Aluminum, calcium, and silicon show significant concentration increases in the fabricated pellets. The aluminum contamination is expected because Al₂O₃ balls were used in the milling operation. The calcium and silicon increases cannot be explained. The silicon contamination is of potential importance because of reported⁸ deleterious effects on densification.

TABLE IV. Plutonium Concentration of Mixed Oxides⁴

Pu (0.1336 nominal), g/g	Pu (0.2666 nominal), g/g
0.1364 ^a	0.2747
0.1377	0.2679
0.1373	
0.1374	

^aTwo samplings were taken from each pellet. Estimated error is $\pm 0.3\%$ of amount present.

TABLE V. Impurity (ppm) Analyses of Mixed Oxides^a

Impurity	U _{0.85} Pu _{0.15}	U _{0.70} Pu _{0.30}	Impurity	U _{0.85} Pu _{0.15}	U _{0.70} Pu _{0.30}
Ag	5	<2.5	K	75	<2.5
Al	480	375	Mg	<50	5
As	<2.5	<2.5	Mn	<2.5	<0.5
Au	<2.5	<2.5	Mo	<2.5	5
B	10	22	Ni	45	120
Ba	10	10	P	100	<500
Bi	<0.05	0.1	Pb	<2.5	<2.5
Ca	<2.5	250	Pd	-	<2.5
Cd	<2.5	<2.5	Sb	<2.5	<2.5
Cr	50	29	Si	>400	>400
Cu	3	1	Sm	<5	10
Fe	95	180	Ti	<2.5	10
Ga	5	20	V	<2.5	<2.5
In	<2.5	<2.5	Zr	<2.5	10

^aAverage from two determinations.

Random continuous scans of 600 μm were run on both compositions to determine inhomogeneity. The scanning rate was 10 $\mu\text{m}/\text{min}$ with a beam size of approximately 1 μm . No variations in uranium or plutonium were noted. X-ray diffraction indicated a high degree of solid solution.

IV. CHARACTERIZATION OF THE FUEL MATERIALS

The fuel pellets received from Dow Chemical Co. were subjected to check analyses by ANL. Checks of chemical analyses consisted of major-element, minor-element, and isotopic content. Each pellet was examined visually for cracks, excessive edge chipping, and hourglassing. Pellets that appeared acceptable on the basis of visual examination were weighed, passed through a 0.349-in.-dia drop-through gauge, and checked for length. These pellets became the population from which fuel columns were assembled. Choice of pellets for a particular column was based upon establishing a limited range of weight-to-length ratios. Each column was identified by assigning it to a specific tube number before loading.

The weight-to-length ratios and standard deviations obtained for 12 columns each of oxide pellets containing 13 wt % Pu, 26 wt % Pu, and depleted UO_2 were 15.69 ± 0.08 , 15.90 ± 0.07 , and 14.92 ± 0.06 g/in., respectively. A tabulation of column lengths and weights, column weight-to-length ratio, and element weight for both prototypic elements and gamma-scan standards is shown in Tables VI and VII.

TABLE VI. Prototype ZPPR Oxide-rod Fuel Elements Produced

Rod Designation	Pellet Column Length, in.	Total Pellet Weight, g	Weight/Length, g/in.	Element Weight, g	ANL Batch No.
<u>13 wt % Pu</u>					
F2	5.726	89.98	15.71	102.31	8-8-1989
F4	5.707	89.68	15.71	102.10	8-8-1989
F6	5.695	89.80	15.77	102.16	8-8-1989
F7	5.702	90.22	15.82	102.59	8-8-1989
F8	5.695	89.73	15.76	102.21	8-8-1989
F9	5.703	88.89	15.58	101.18	8-8-1990
F11	5.705	89.00	15.60	101.20	8-8-1990
F15	5.710	89.15	15.61	101.45	8-8-1990
F18	5.712	89.84	15.73	102.28	8-8-1989
F19	5.706	89.80	15.74	102.19	8-8-1989
F20	5.726	89.41	15.61	101.63	8-8-1990
F22	5.745	89.66	15.61	101.79	8-8-1990
<u>26 wt % Pu</u>					
G12	5.676	89.89	15.85	102.07	8-8-1987
G24	5.670	89.83	15.84	101.89	8-8-1987
G27	5.679	89.77	15.80	102.01	8-8-1987
G29	5.670	90.19	15.89	102.55	8-8-1987
G31	5.676	90.19	15.89	102.61	8-8-1987
G33	5.708	91.10	15.96	103.48	8-8-1987
G34	5.715	91.36	15.98	103.68	8-8-1987
G35	5.698	91.36	16.03	103.88	8-8-1987
G36	5.680	90.11	15.86	102.43	8-8-1987
G39	5.672	89.83	15.83	102.09	8-8-1987
G43	5.671	90.89	16.02	103.42	8-8-1987
G44	5.677	90.00	15.85	102.12	8-8-1987
<u>Depleted Uranium Dioxide</u>					
D51	5.755	86.25	14.89	98.69	6-6-686
D53	5.755	86.22	14.98	98.23	6-6-686
D54	5.755	86.26	14.98	98.47	6-6-686
D55	5.745	85.30	14.84	97.67	6-6-686
D56	5.755	85.97	14.93	98.21	6-6-686
D57	5.755	86.27	14.99	98.60	6-6-686
D60	5.755	86.24	14.98	98.50	6-6-686
D62	5.746	85.23	14.83	97.51	6-6-686
D67	5.755	85.82	14.91	98.14	6-6-686
D68	5.755	86.17	14.97	98.19	6-6-686
D70	5.747	85.34	14.85	97.72	6-6-686
D71	5.746	85.29	14.84	97.28	6-6-686

TABLE VII. ZPPR Oxide-rod Gamma-scan Standards

Rod Designation	Nominal Composition, wt %	Pellet Column Length, in.	Total Pellet Weight, g	Weight/Length, g/in.	Element Weight, g	ANL Batch No.
101	10-Pu	5.736	89.51	15.60	101.84	8-8-2124
102	16-Pu	5.736	88.13	15.86	100.47	8-8-2128
104	16-Pu	5.737	87.22	15.20	99.33	8-8-2128
87	21-Pu	5.723	89.71	15.67	101.89	8-8-2143
88	21-Pu	5.712	89.72	15.70	101.96	8-8-2143
89	28-Pu	5.643	89.72	15.90	102.51	8-8-2151
90	28-Pu	5.646	89.75	15.89	102.45	8-8-2151
77	6-235U	5.689	88.14	15.49	100.79	36-2-482
92	7-235U	5.738	84.08	14.65	96.37	8-8-2138
93	7-235U	5.747	84.17	14.64	96.08	8-8-2138
94	14-235U	5.738	82.49	14.37	94.99	8-8-2139
95	14-235U	5.738	82.87	14.39	95.09	8-8-2139
96	21-235U	5.739	84.81	14.77	97.18	8-8-2140
98	21-235U	5.739	84.67	14.73	97.17	8-8-2140
99	37-235U	5.738	85.33	14.87	97.79	8-8-2141
100	37-235U	5.738	85.41	14.88	97.99	8-8-2141

V. HARDWARE ASSEMBLY AND INSPECTION

A. Forming

The Type 304L stainless steel hardware, consisting of two end plugs and a tube, was machined to dimensions specified in Figs. 3 and 4. One end of the tube was expanded over a 0.363-dia mandrel held on a common centerline with a lathe collet that held the tube. The depth of expansion was controlled by a stop located on the mandrel. The tube was expanded to accommodate a loading funnel that had the same inside diameter as the jacket tubing. Each tube was identified by electroetching a number 0.5 in. from one end. The end plugs and tube were washed in acetone and then rinsed in ethyl alcohol. Pieces were then placed in a circulating dry-air furnace at 100°C for 1 hr before transferring to the welding hoodline. The minimum of handling time was an important consideration in choosing the fabrication scheme outlined in Figs. 11a, b, and c.

B. Welding

The unexpanded end of the jacket tube was clamped in a split copper chill and positioned so that 0.018 in. of the end to be welded extended beyond the chill. The end cap was inserted and the assembly placed in a lathe collet held in a vertical position. A second copper chill was placed in the recess of the end plug. This combination of chills was required to hold the weld bead in an upright position until solidification occurred, producing full thickness of the tube wall at the junction of the end plug and jacket tube. After welding, the jacket tubes were inspected for ovality, straightness, and weld-bead protrusion by insertion into a 0.380-in.-ID drop-through gauge to the expanded portion.

A 1% thoriated tungsten electrode held at 45° and positioned 0.031 in. above the end cap was used during welding. Figure 12 shows the welding setup, including the chilling arrangements. The TIG weld was made by using a constant 22 rpm through 370° of rotation before starting current decay. A low power of 26 A dc at 12-14 V with a flow of 25 cfm helium and 10 cfm argon was used to make the weld shown in Fig. 5. The bottom end-cap weld was tested for integrity by means of helium mass-spectrometer leak detection. The method consisted of pulling a vacuum on the inside of the jacket tube while the outside of the tube was exposed to helium. An assembly showing a leak indication above background was rejected.

VI. LOADING, ASSEMBLY, AND INSPECTION OF THE FUEL ELEMENT

To prevent contamination in the final weld closure, a special loading funnel was inserted in the expanded portion of the jacket tube; the small lip on the funnel extending into the inside diameter of the jacket tube below the thickness of the end plug. A 0.010-in.-dia Nichrome wire was stretched along the length of the funnel and the assembly. Shrinkable plastic tubing was slipped over this assembly and the entire assembly was placed in a drying oven at 160°C for 10 min. The components are shown, in Fig. 13, before assembly, after assembly, and after

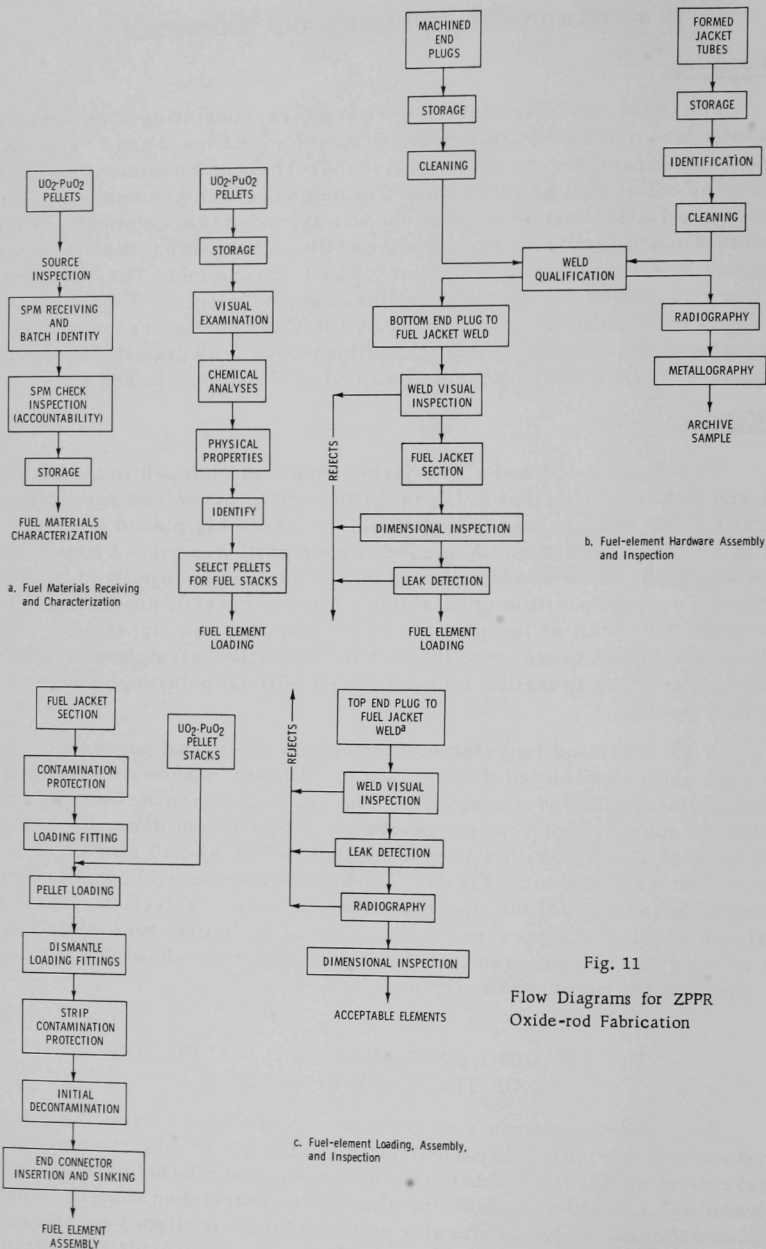


Fig. 11
Flow Diagrams for ZPPR
Oxide-rod Fabrication



Fig. 12

ZPPR Oxide-rod Welding Arrangement.
ANL Neg. No. 350-1086.



Fig. 13. Exploded and Cutaway Views of ZPPR Oxide-rod Elements. ANL Neg. No. 350-1073.

After the shrinkable tubing was trimmed to length, the assembly was secured to a "cow" by means of compressive hose clamps to insure a helium leak-tight seal between the jacket and the fitting (Fig. 14). The "cow" consists of a vinyl pouch attached to a gloveport in the loading box. The vinyl pouch has four polyvinylchloride fittings, dielectrically attached. A vinyl membrane over the face of the fitting was slit to expose the funnel end of

the tube to the helium atmosphere in the hood. A column of pellets that had been weighed and measured for length and diameter were loaded into each jacket. Each pellet was bottomed into the jacket with a push rod. After the last pellet was inserted, the vinyl pouch was dielectrically sealed and cut free. The pellet loading is shown in Fig. 15.



Fig. 14. Out-of-glovebox View of "Cow" Loading Fixture. ANL Neg. No. 350-1080.



Fig. 15
ZPPR Oxide-pellet Loading through Vinyl
"Cow." ANL Neg. No. 350-1079.

The loaded element with a portion of the "cow" attached was transferred to an annex box of the welding hood. The vinyl cow was removed from the element, and the exposed end of the loading funnel was cleaned of loose contamination with paper wipes and cotton swabs moistened with ethyl alcohol. The shrinkable tubing was removed by pulling on the Nichrome wire, which acts as a zipper. The loading funnel was removed and the inside of the fuel jacket checked for contamination, being cleaned if necessary. The cumulative tolerance on the length of the pellet stack could not be determined until the pellets were settled in the jacket tube. A micrometer reading was taken from the end of the jacket tube to the top of the first pellet to determine the length of the spacer required to give the required 0.000-0.010-in. end cap-to-spacer clearance. A spacer of the proper thickness was cut, inserted, and checked again with a micrometer before insertion of the endplug. The expanded end of the jacket tube was reduced to its original diameter by using a special collet operated by an adjustable over-center cam, as shown in Fig. 16. A split brass collet was inserted

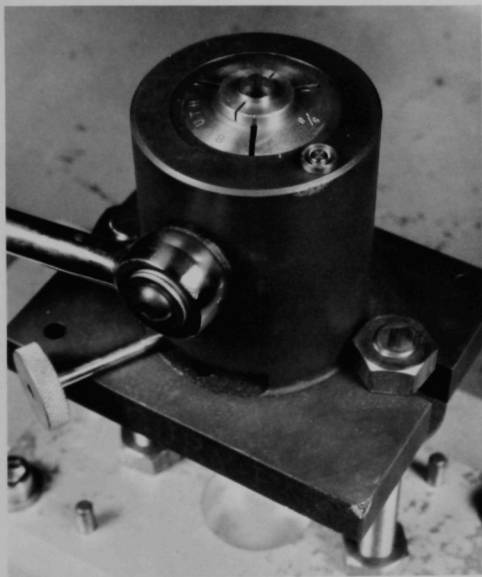


Fig. 16. ZPPR Oxide-rod Sinking with Over-center Cam. ANL Neg. No. 350-1085.

in a standard lathe collet, and the tube was compressed in four stages. The jacket was rotated between each stage to eliminate marking the tube at the edges of the collet. The top edge of the jacket was checked for loose contamination before the insertion of the end plug. The final end plug was tapped into position before transferring the assembly to the welding hood. The final end closure was made with the same procedure outlined in Part V.B of this report.

The completed fuel-element assembly was checked for loose and fixed contamination before checking for leaks with a helium mass spectrometer. Acceptable contamination levels were <400 dpm fixed and <10 dpm loose alpha counts. The final end closure was made in an atmosphere that contained helium. The elements were placed one at a time

in a fixture that reduced the pressure and used the entrapped helium as the leak indicator. As in the case of leak detection of the bottom end plug any leak indication above background was cause for rejection. All elements were radiographed to check welds, and position of the fuel and end spacer.

Tables VI and VII present a record of the 52 completed elements fabricated for reactor physics experiments and gamma-scan standards for evaluating ZPPR oxide rods produced by a commercial vendor.

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REFERENCES

1. Caldwell, C. S., and Puechl, K. H., *Plutonium-Uranium Dioxide Powder and Pellet Fuel Manufacture*, Nuclear Metallurgy, AIME 13, 174-194 (1968).
2. Ruther, W. E., and Greenberg, S., *Corrosion of Steels and Nickel Alloys in Superheated Steam*, J. Electrochem. Soc. 111(10), 1116-1121 (Oct 1964).
3. Gulbransen, E. A., and Andrew, K. F., *Oxidation Studies on 304 Stainless Steel*, J. Electrochem. Soc. 109(7), 560-564 (July 1962).
4. Harvey, M. R., Teter, A. R., and Leggett, R. L., *Fabrication of Oxide Nuclear Fuel Pellets*, The Dow Chemical Co., Rocky Flats Division, Golden, Colorado, RFP-1255 (March 1969).
5. Stakebake, J. L., The Dow Chemical Co., Rocky Flats Division, Golden, Colorado, private communication.
6. Lyon, W. L., *The Measurement of Oxygen to Metal Ratio in Solid Solutions of Uranium and Plutonium Dioxides*, GEAP-4271 (1963).
7. Runfors, U., and Kiessling, R., *Sintering of Uranium Dioxide*, TID-7546, Book 2, 402-413 (1958).
8. Caldwell, C. S., and Thomas, I. D., *Plutonium Uranium Mixed Oxide Preparation and Fabrication Experience*, Nucl. Eng., Part XVIII, 63(80), 147-155 (1966).

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